Ultrafast dynamics of quantum many-body systems including dynamical screening and strong coupling

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Strongly Coupled Coulomb Systems

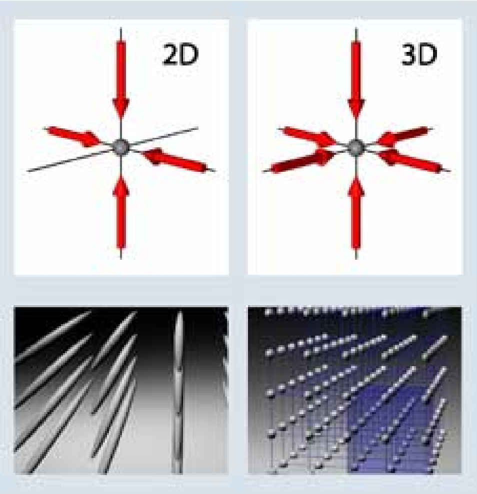
Görlitz, July 2022

pdf at http://www.theo-physik.uni-kiel.de/bonitz/talks.html

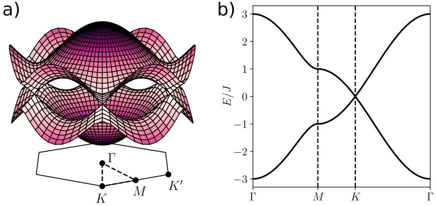
Finite correlated quantum systems

Fermionic atoms in optical lattices

tunable lattice depth and interaction



Graphene: high mobility, no bandgap



Graphene nanoribbons:

finite tunable bandgap

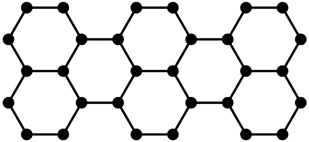
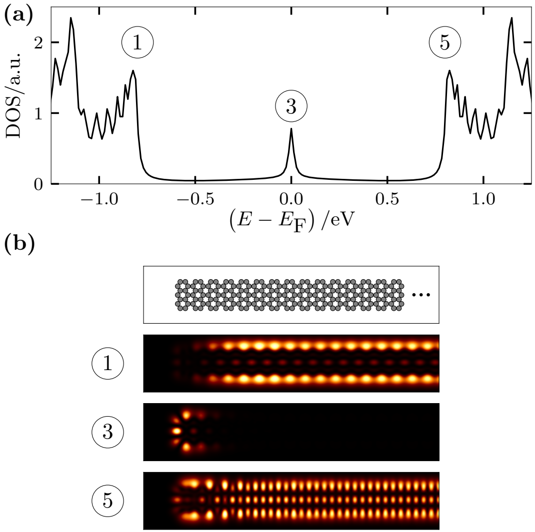


Fig.: M. Greiner (Harvard)

GNR: spatially localized spectral contributions[1]

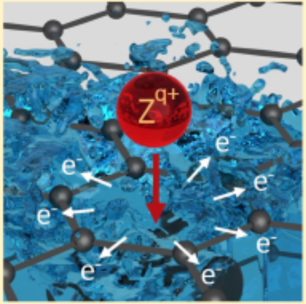


- top: total density of states (DOS)
- DOS size and shape dependent
- many degrees of freedom: combination of materials, multiple layers
- importance of e-e interactions
- what will happen in nonequilibrium, upon external excitation (e.g. by lasers)?

[1] 7 armchair GNR of 504 atoms, GW-NEGF ground state simulation, J.-P. Joost, A.-P. Jauho, and M. Bonitz, Nano Letters **19**, 9045 (2019)

Ion stopping in 2D quantum materials[2]

ABSTRACT: Low-energy electrons (LEEs) are of great relevance for ion-induced radiation damage in cells and genes. We show that charge exchange of ions leads to LEE emission upon impact on condensed matter. By using a graphene monolayer as a simple model system for condensed organic matter and utilizing slow highly charged ions (HCIs) as projectiles, we highlight the importance of charge exchange alone for LEE emission. We find a large number of ejected electrons resulting from individual ion impacts (up to 80 electrons/ion for Xe^{40+}). More than 90% of emitted electrons have energies well below 15 eV. This "splash" of low-energy electrons is interpreted as the consequence of ion deexcitation via an interatomic Coulombic decay (ICD) process.

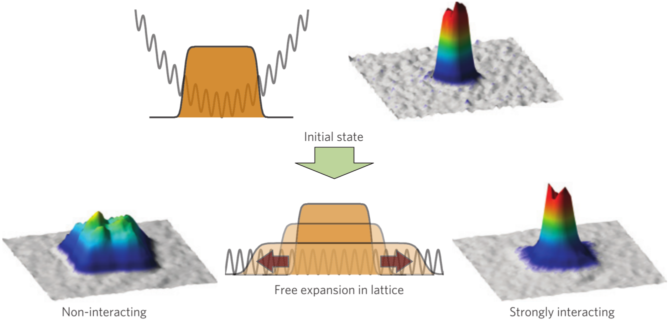


- traversal of energetic ions through graphene-type monolayers: very strong localized excitation
- complex processes: energy deposition, charge exchange
- emission of low-energy electrons, strong dependence on target properties
- ultrafast electronic processes (1...5 fs)

[2]Schwestka *et al.*, J. Phys. Chem. Lett. **10**, 4805 (2019),
A. Niggas, ... R. Wilhelm, ... K. Balzer, N. Schluenzen, ...M. Bonitz, PRL 2022, in press

Cold atoms in optical lattices: quench induced dynamics

- Prepare cold atoms at a given coupling strength U
- "Instantly" change the system parameters
- Observe the many-particle dynamics
- Question: how does the interaction strength influence the dynamics?



Diffusion of cold fermionic atoms following a confinement quench

[1] Schneider et al., Nature Phys. (2012).

Time-dependent Schrödinger equation. Scaling bottleneck

- time-dependent many-electron Hamiltonian

$$H(t) = \underbrace{\sum_{i=1}^{N} h(\boldsymbol{r}_i, t)}_{\text{one-body operators}} + \underbrace{\frac{1}{2}\sum_{i\neq j}^{N} W(\boldsymbol{r}_i, \boldsymbol{r}_j)}_{\textbf{pair-wise}\text{ interactions}}$$

- time-dependent Schrödinger equation (TDSE)

$$i\partial_t \Psi(\boldsymbol{r}_i, \dots, \boldsymbol{r}_N; t) = H(t)\Psi(\boldsymbol{r}_i, \dots, \boldsymbol{r}_N; t) \quad \overset{\text{direct solution}}{\underset{\text{exponential scaling of numerical effort}}{\not\Longrightarrow}} \quad \Psi(\boldsymbol{r}_i, \dots, \boldsymbol{r}_N; t)$$

- **solutions to overcome exponential scaling:**
  1. approximations to TDSE: TD-RASCI, TD-CASSCF, truncated CC, TD-R-matrix etc. D. Hochstuhl and M. Bonitz, PRA (2012) and EJP-ST (2014)
  2. propagation of simpler observables: density (TDDFT), distribution function (Kinetic theory), correlation functions etc.

Electron dynamics in plasmas with kinetic equations

- **Boltzmann's kinetic equation for the phase space distribution** $f(\mathbf{r}_1, \mathbf{p}_1, t)$

$$\frac{\partial f}{\partial t} + \frac{\mathbf{p}_1}{m} \cdot \nabla f + \mathbf{F}^{\text{tot}} \cdot \frac{\partial f}{\partial \mathbf{p}_1} = \int dp_2 dp_1' dp_2' \, \sigma(p_1, p_2; p_1', p_2') \left\{ f_1' f_2' - f_1 f_2 \right\} \Big|_t = I(p_1, t)$$

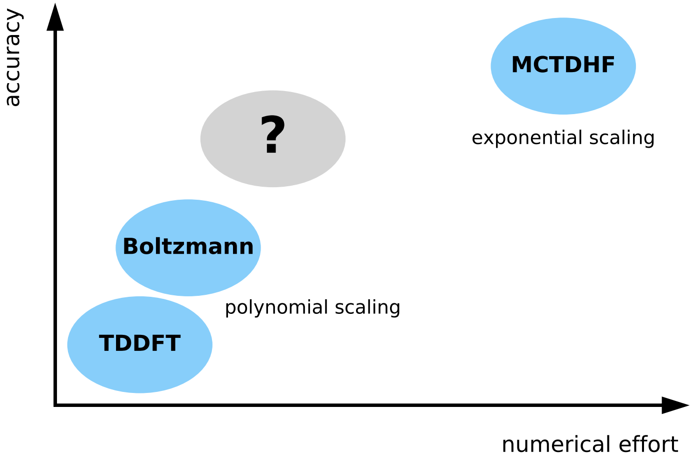
- I : two-particle scattering effects, modified by surrounding medium (e.g. screening)
- static screening: Landau; dynamic screening: Balescu-Lenard equation.

$$\sigma^{\text{BL}} \sim \left| \frac{V(p_1 - p_1')}{\epsilon\left(p_1 - p_1', E_{p_1} - E_{p_1'}\right)} \right|^2 \delta(p_1 + p_2 - p_1' - p_2') \, \delta(E_{p_1} + E_{p_2} - E_{p_1'} - E_{p_2'})$$

- **Problems of the Boltzmann and Balescu equations:**[3]
  1. neglect of strong coupling/multiple scattering effects (T-matrix diagrams)
  2. no total energy conservation
  3. not applicable to femtosecond time scales (no correlation buildup)

[3] for details, see M. Bonitz, *Quantum Kinetic Theory*, 2nd ed., Springer 2016

Scaling of quantum many-body methods



*MCTDHF and other wavefunction-based methods

? Can one achieve sufficient accuracy (including e-e correlations) at non-exponential cost?

2nd quantization

- Fock space $\mathcal{F} \ni |n_1, n_2 \ldots\rangle$, $\mathcal{F} = \bigoplus_{N_0 \in \mathbb{N}} \mathcal{F}^{N_0}$, $\mathcal{F}^{N_0} \subset \mathcal{H}^{N_0}$
- $\hat{c}_i$,$\hat{c}_i^\dagger$ creates/annihilates a particle in single-particle orbital ϕ_i
- spin accounted for by canonical (anti-)commutator relations
  $\left[\hat{c}_i^{(\dagger)}, \hat{c}_j^{(\dagger)}\right]_\mp = 0, \quad \left[\hat{c}_i, \hat{c}_j^\dagger\right]_\mp = \delta_{i,j}$
- Hamiltonian: $\hat{H}(t) = \underbrace{\sum_{k,m} h^0_{km} \hat{c}_k^\dagger \hat{c}_m}_{\hat{H}_0} + \underbrace{\frac{1}{2} \sum_{k,l,m,n} w_{klmn} \hat{c}_k^\dagger \hat{c}_l^\dagger \hat{c}_n \hat{c}_m}_{\hat{W}} + \hat{F}(t)$

Particle interaction w_{klmn}

- Coulomb interaction
- electronic correlations

Time-dependent excitation $\hat{F}(t)$

- single-particle type
- em field, quench, particles

Nonequilibrium Green Functions (NEGF)

two times $z, z' \in \mathcal{C}$ ("Keldysh contour"), arbitrary one-particle basis $|\phi_i\rangle$

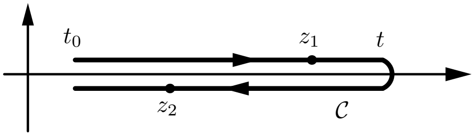
$$\boxed{G_{ij}(z,z') = \frac{\mathrm{i}}{\hbar}\left\langle \hat{T}_{\mathcal{C}}\hat{c}_i(z)\hat{c}_j^\dagger(z')\right\rangle}$$

average with $\hat{\rho}_N$

pure or mixed state

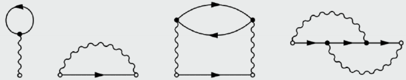
Keldysh–Kadanoff–Baym equations (KBE) on $\mathcal{C}$ (2×2 matrix):

$$\sum_k \left\{ \mathrm{i}\hbar\frac{\partial}{\partial z}\delta_{ik} - h_{ik}(z)\right\} G_{kj}(z,z') = \delta_{\mathcal{C}}(z,z')\delta_{ij} - \mathrm{i}\hbar\sum_{klm}\int_{\mathcal{C}} \mathrm{d}\bar{z}\, w_{iklm}(z^+,\bar{z})G^{(2)}_{lmjk}(z,\bar{z};z',\bar{z}^+)$$



KBE: first equation of Martin–Schwinger hierarchy for $G, G^{(2)} \ldots G^{(n)}$

- $\int_{\mathcal{C}} wG^{(2)} \to \int_{\mathcal{C}} \Sigma G$, Selfenergy
- Nonequilibrium Diagram technique
  Example: Hartree–Fock + Second Born selfenergy

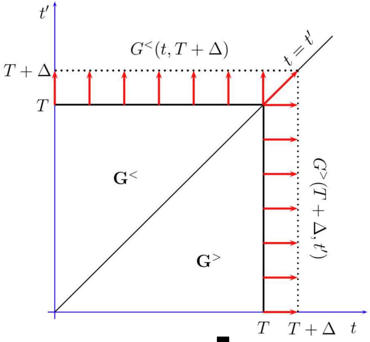


Real-Time Keldysh–Kadanoff–Baym Equations (KBE)

- Correlation functions $G^{\gtrless}$ obey real-time KBE

$$\sum_l \left[\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} \delta_{i,l} - h^{\text{eff}}_{il}(t) \right] G^{>}_{lj}(t,t') = I^{(1),>}_{ij}(t,t') \,,$$

$$\sum_l G^{<}_{il}(t,t') \left[-\mathrm{i}\hbar \frac{\overleftarrow{\mathrm{d}}}{\mathrm{d}t'} \delta_{l,j} - h^{\text{eff}}_{lj}(t') \right] = I^{(2),<}_{ij}(t,t') \,,$$



with the effective single-particle **Hartree–Fock Hamiltonian**

$$h^{\text{eff}}_{ij}(t) = h^0_{ij} \pm \mathrm{i}\hbar \sum_{kl} w^{\pm}_{ikjl} G^{<}_{lk}(t)$$

and the collision integrals

$$I^{(1),>}_{ij}(t,t') := \sum_l \int_{t_\text{s}}^{\infty} \mathrm{d}\bar{t} \left\{ \Sigma^{\text{R}}_{il}(t,\bar{t}) G^{>}_{lj}(\bar{t},t') + \Sigma^{>}_{il}(t,\bar{t}) G^{\text{A}}_{lj}(\bar{t},t') \right\} ,$$

$$I^{(2),<}_{ij}(t,t') := \sum_l \int_{t_\text{s}}^{\infty} \mathrm{d}\bar{t} \left\{ G^{\text{R}}_{il}(t,\bar{t}) \Sigma^{<}_{lj}(\bar{t},t') + G^{<}_{il}(t,\bar{t}) \Sigma^{\text{A}}_{lj}(\bar{t},t') \right\} .$$

→ $\mathcal{O}(N_\text{t}^3)$

- two-time structure contains spectral information
- numerically demanding due to cubic scaling with number of time steps N_t

Selfenergy Approximations[4]

Choice depends on coupling strength, density (filling)

Hartree–Fock (HF, mean field): $\sim w^1$

Second Born (2B): $\sim w^2$

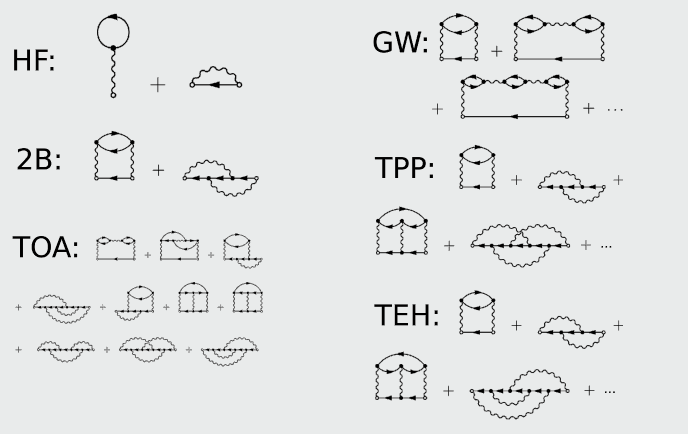
$\boldsymbol{GW}$: ∞ bubble summation,
dynamical screening effects

particle-particle T-matrix (TPP):
∞ ladder sum in pp channel

particle-hole T-matrix (TPH/TEH):
∞ ladder sum in ph channel

3rd order approx. (TOA): $\sim w^3$

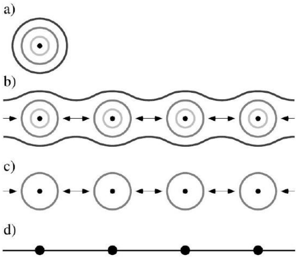
dynamically screened ladder (DSL):
$\sim \mathrm{2B} + GW + \mathrm{TPP} + \mathrm{TPH}$



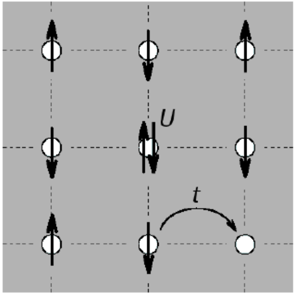
[4] Conserving approximations, nonequilibrium $\Sigma(t, t')$, applies for ultra-short to long times
Review: Schlünzen *et al.*, J. Phys. Cond. Matt. **32**, 103001 (2020)

Testing various selfenergies: the Hubbard model

- Simple, but versatile model for strongly correlated solid state systems
- Suitable for single band, small bandwidth



$\Rightarrow$



$$\hat{H}(t) = J \sum_{ij,\,\alpha} h_{ij}\, \hat{c}^{\dagger}_{i\alpha} \hat{c}_{j\alpha} + U \sum_{i} \hat{c}^{\dagger}_{i\uparrow} \hat{c}_{i\uparrow} \hat{c}^{\dagger}_{i\downarrow} \hat{c}_{i\downarrow} + \sum_{ij,\alpha\beta} f_{ij,\alpha\beta}(t)\, \hat{c}^{\dagger}_{i\alpha} \hat{c}_{j\beta}$$

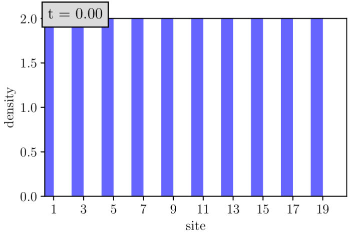
$h_{ij} = -\delta_{\langle i,\,j\rangle}$ and $\delta_{\langle i,\,j\rangle} = 1$, if (i, j) is nearest neighbor, $\delta_{\langle i,\,j\rangle} = 0$ otherwise

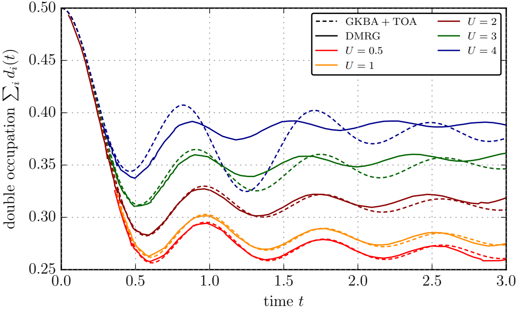
use $J = 1$, on-site repulsion $(U > 0)$ or attraction $(U < 0)$, tunable interaction strength

Benchmarks of NEGF against DMRG (1D)[5]

Initial state:

charge density wave

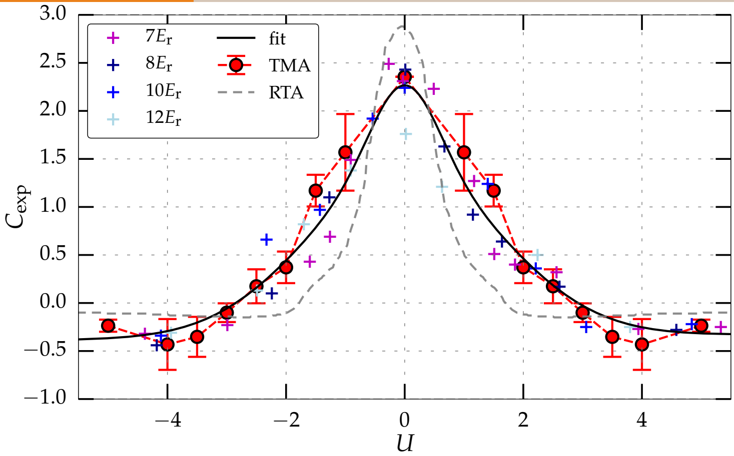




- sensitive observable: total double occupation
- good quality transients NEGF up to $U \simeq$ bandwidth
- Accurate long-time behavior of GKBA+T-matrix (not shown)

[5] N. Schlünzen, J.-P. Joost, F. Heidrich-Meisner, and M. Bonitz, Phys. Rev. B **95**, 165139 (2017)

Core expansion velocity: NEGF result[6] vs. experiment and RTA[7]



- Many-fermion expansion following sudden removal of confinement: interaction effects
- agreement with measurements for the *final stage* of the dynamics
- in addition: NEGF predict early stages, correlation dynamics etc.

[6] N. Schlünzen, S. Hermanns, M. Bonitz, and C. Verdozzi, Phys. Rev. B **93**, 035107 (2016)

[7] U. Schneider *et al.*, Nature Physics **8**, 213-218 (2012)

Acceleration: Generalized Kadanoff–Baym Ansatz (GKBA)[8]

- full propagation on the time diagonal ($I := I^{(1),<}$):

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}G^{<}_{ij}(t) = \left[h^{\mathrm{HF}}, G^{<}\right]_{ij}(t) + \left[I + I^{\dagger}\right]_{ij}(t)$$

- reconstruct off-diagonal NEGF from time diagonal:

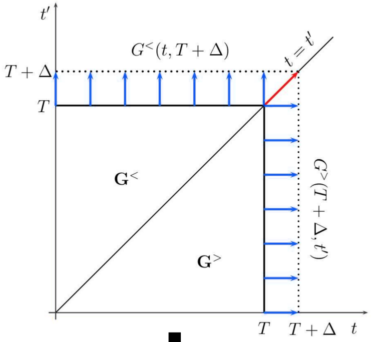
$$G^{\gtrless}_{ij}(t,t') = \pm\left[G^{\mathrm{R}}_{ik}(t,t')\rho^{\gtrless}_{kj}(t') - \rho^{\gtrless}_{ik}(t)G^{\mathrm{A}}_{kj}(t,t')\right]$$

with $\quad \rho^{\gtrless}_{ij}(t) = \pm\mathrm{i}\hbar G^{\gtrless}_{ij}(t,t)$

- HF-GKBA: use Hartree–Fock propagators for $G^{\mathrm{R/A}}_{ij}$

$$G^{\mathrm{R/A}}_{ij}(t,t') = \mp\mathrm{i}\Theta_{\mathcal{C}}\left(\pm[t-t']\right)\exp\left(-\frac{\mathrm{i}}{\hbar}\int_{t'}^{t}\mathrm{d}\bar{t}\, h_{\mathrm{HF}}(\bar{t})\right)\Bigg|_{ij}$$

- conserves total energy

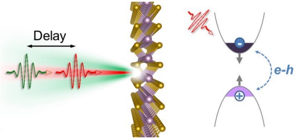


$$\mathcal{O}(N_{\mathrm{t}}^2)$$

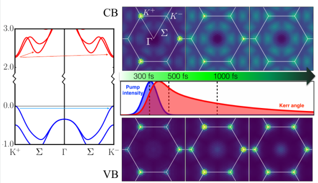
[6] P. Lipavský, V. Špička, and B. Velický, Phys. Rev. B **34**, 6933 (1986);
K. Balzer and M. Bonitz, Lecture Notes in Physics **867** (2013)

GKBA results for materials, plasmas

2D Layered Materials

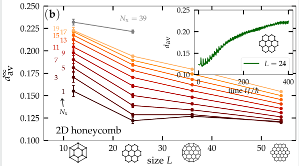


E. A. Pogna *et al.*,
ACS Nano **10**, 1182 (2016)



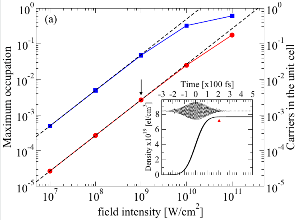
A. Molina-Sánchez *et al.*,
Nano Lett. **17**, 4549 (2017)

Ion Stopping in Hexagonal Lattices



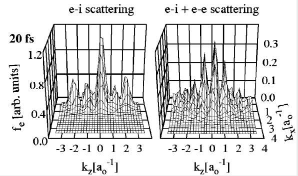
K. Balzer *et al.*,
PRL **121**, 267602 (2018)

Semiconductors



D. Sangalli *et al.*,
PRB **93**, 195205 (2016)

Laser-Induced Heating of Dense Plasmas



H. Haberland *et al.*,

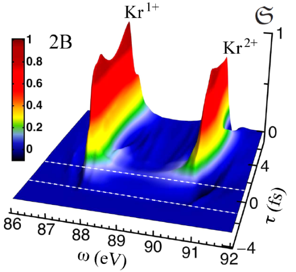
PRE **64**, 026405 (2001)
gauge-invariant

multi-photon absorption

inv bremsstrahlung heating

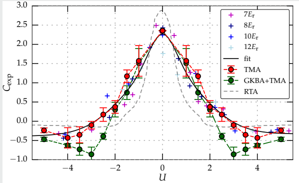
GKBA results for atoms and molecules

Atoms



E. Perfetto *et al.*,
PRA **92**, 033419 (2015)

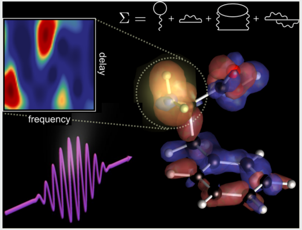
Cold Atoms in Optical Lattices



N. Schlünzen, M. Bonitz,
CPP **56**, 5 (2016)

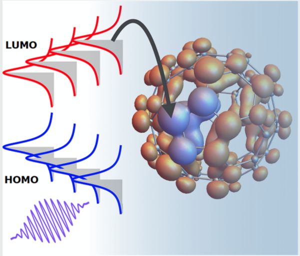
Σ beyond 2nd order

Biologically Relevant Molecules



E. Perfetto *et al.*,
JCPL **9**, 1353 (2018)

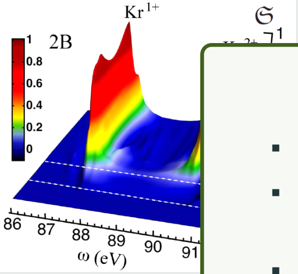
Carbon Allotropes



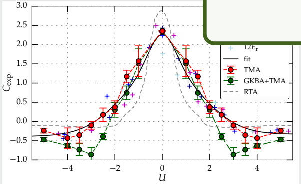
E. V. Boström *et al.*,
Nano Lett. **18**, 785 (2018)

GKBA results for atoms and molecules

Atoms



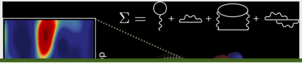
Cold Atoms in Optica



N. Schlünzen, M. Bonitz, CPP **56**, 5 (2016)

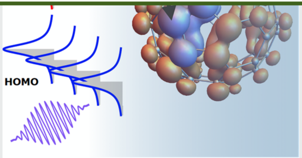
Σ beyond 2nd order

Biologically Relevant Molecules



etto *et al.*,

, 1353 (2018)



E. V. Boström *et al.*, Nano Lett. **18**, 785 (2018)

but

- improvement to N_t^2 scaling only possible for 2B selfenergy
- typical systems with small $N_b \sim 10$–100 but large $N_t \sim 1000$–10000
- still huge numerical disadvantage compared to other linearly scaling methods (TD-DMRG, TDDFT, TDSE)

Is $\mathcal{O}(N_t^1)$ scaling possible?

Reformulating the GKBA

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \int_{t_0}^{t} \mathrm{d}\bar{t} \left[\Sigma^{>}_{ik}(t,\bar{t}) G^{<}_{kj}(\bar{t},t) - \Sigma^{<}_{ik}(t,\bar{t}) G^{>}_{kj}(\bar{t},t)\right]$$

time integral → $\int_{t_0}^{t} \mathrm{d}\bar{t}$

off-diagonal functions → $(t,\bar{t})$, $(\bar{t},t)$, $(t,\bar{t})$, $(\bar{t},t)$

Reformulating the GKBA

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time integral

off-diagonal functions

Idea: solve differential equation for $\mathcal{G}$ instead of time integral for I

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \int_{t_0}^{t} \mathrm{d}\bar{t} \left[\Sigma^{>}_{ik}(t,\bar{t}) G^{<}_{kj}(\bar{t},t) - \Sigma^{<}_{ik}(t,\bar{t}) G^{>}_{kj}(\bar{t},t)\right] =: \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t) \mathcal{G}_{lpjk}(t)$$

time integral — off-diagonal functions — **Idea:** solve differential equation for $\mathcal{G}$ instead of time integral for I

- example for 2B selfenergy[9]

$$\Sigma^{\gtrless}_{ij}(t,t') = \pm (\mathrm{i}\hbar)^2 \sum_{klpqrs} w_{iklp}(t)\, w^{\pm}_{qrjs}(t')\, G^{\gtrless}_{lq}(t,t')\, G^{\gtrless}_{pr}(t,t')\, G^{\lessgtr}_{sk}(t',t)$$

- respective $\mathcal{G}$ can be identified as

$$\mathcal{G}_{ijkl}(t) = \mathrm{i}\hbar \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, w^{\pm}_{pqrs}(\bar{t}) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,\bar{t}) \mathcal{G}^{\mathrm{H},<}_{rskl}(\bar{t},t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,\bar{t}) \mathcal{G}^{\mathrm{H},>}_{rskl}(\bar{t},t)\right]$$

with the two-particle Hartree Green function

$$\mathcal{G}^{\mathrm{H},\gtrless}_{ijkl}(t,t') := G^{\gtrless}_{ik}(t,t') G^{\gtrless}_{jl}(t,t')$$

[9] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)

Reformulating the GKBA

- quadratic/cubic scaling is caused by the structure of the collision integral

$$I_{ij}(t) = \sum_k \int_{t_0}^{t} \mathrm{d}\bar{t} \left[\Sigma^{>}_{ik}(t,\bar{t}) G^{<}_{kj}(\bar{t},t) - \Sigma^{<}_{ik}(t,\bar{t}) G^{>}_{kj}(\bar{t},t)\right] =: \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t) \mathcal{G}_{lpjk}(t)$$

time integral — off-diagonal functions — **Idea:** solve differential equation for $\mathcal{G}$ instead of time integral for I

- example for 2B selfenergy[9]

$$\Sigma^{\gtrless}_{ij}\left(t,t'\right) = \pm\left(\mathrm{i}\hbar\right)^2 \sum_{klpqrs} w_{iklp}\left(t\right) w^{\pm}_{qrjs}\left(t'\right) G^{\gtrless}_{lq}\left(t,t'\right) G^{\gtrless}_{pr}\left(t,t'\right) G^{\lessgtr}_{sk}\left(t',t\right)$$

- respective $\mathcal{G}$ can be identified as

$$\mathcal{G}_{ijkl}(t) = \mathrm{i}\hbar \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, w^{\pm}_{pqrs}\left(\bar{t}\right) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,\bar{t}) \mathcal{G}^{\mathrm{H},<}_{rskl}(\bar{t},t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,\bar{t}) \mathcal{G}^{\mathrm{H},>}_{rskl}(\bar{t},t)\right]$$

with the two-particle Hartree Green function

$$\mathcal{G}^{\mathrm{H},\gtrless}_{ijkl}(t,t') := G^{\gtrless}_{ik}(t,t') G^{\gtrless}_{jl}(t,t')$$

[9] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)

Reformulating the GKBA

- two-particle $\mathcal{G}$ in GKBA

$$\mathcal{G}_{ijkl}(t) = (\mathrm{i}\hbar)^3 \sum_{pqrs} \int_{t_0}^{t} \mathrm{d}\bar{t}\, \mathcal{U}^{(2)}_{ijpq}(t,\bar{t}) \Psi^{\pm}_{pqrs}(\bar{t}) \mathcal{U}^{(2)}_{rskl}(\bar{t},t)$$

with the single-time source term (which no longer depends on the outer time)

$$\Psi^{\pm}_{ijkl}(t) = (\mathrm{i}\hbar)^2 \sum_{pqrs} w^{\pm}_{pqrs}(t) \left[\mathcal{G}^{\mathrm{H},>}_{ijpq}(t,t) \mathcal{G}^{\mathrm{H},<}_{rskl}(t,t) - \mathcal{G}^{\mathrm{H},<}_{ijpq}(t,t) \mathcal{G}^{\mathrm{H},>}_{rskl}(t,t) \right]$$

and the two-particle Hartree–Fock time-evolution operators obeying Schrödinger-type EOMs

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[\mathcal{U}^{(2)}_{ijkl}(t,\bar{t}) \right] = \frac{1}{\mathrm{i}\hbar} \sum_{pq} h^{(2),\mathrm{HF}}_{ijpq}(t) \mathcal{U}^{(2)}_{pqkl}(t,\bar{t})$$

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[\mathcal{U}^{(2)}_{ijkl}(\bar{t},t) \right] = -\frac{1}{\mathrm{i}\hbar} \sum_{pq} \mathcal{U}^{(2)}_{ijpq}(\bar{t},t) h^{(2),\mathrm{HF}}_{pqkl}(t)$$

with the effective two-particle Hamiltonian

$$h^{(2),\mathrm{HF}}_{ijkl}(t) = \delta_{jl} h^{\mathrm{HF}}_{ik}(t) + \delta_{ik} h^{\mathrm{HF}}_{jl}(t)$$

The G1–G2 Scheme

- full propagation on the time diagonal as for ordinary HF-GKBA:

$$\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} G^{<}_{ij}(t) = \left[h^{\mathrm{HF}}, G^{<}\right]_{ij}(t) + \left[I + I^{\dagger}\right]_{ij}(t)$$

- but collision integral defined by correlated two-particle Green function

$$I_{ij}(t) = \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t) \mathcal{G}_{lpjk}(t)$$

- which obeys an ordinary differential equation[10]

$$\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} \mathcal{G}_{ijkl}(t) = \left[h^{(2),\mathrm{HF}}, \mathcal{G}\right]_{ijkl}(t) + \Psi^{\pm}_{ijkl}(t)$$

$$\rightarrow \mathcal{O}(N_{\mathrm{t}}^{1})$$

[10] two-particle commutator: $[A, B]_{ijkl}(t) = \sum_{pq} \left[A_{ijpq}(t) B_{pqkl}(t) - B_{ijpq}(t) A_{pqkl}(t)\right]$

The G1–G2 Scheme

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- full propagation on the time diagonal as for ordinary HF-GKBA:

$$\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} G^<_{ij}(t) = \left[h^{\mathrm{HF}}, G^<\right]_{ij}(t) + \left[I + I^\dagger\right]_{ij}(t)$$

- but collision integral defined by correlated two-particle Green function

$$I_{ij}(t) = \pm \mathrm{i}\hbar \sum_{klp} w_{iklp}(t) \mathcal{G}_{lpjk}(t)$$

- which obeys an ordinary differential equation[10]

$$\mathrm{i}\hbar \frac{\mathrm{d}}{\mathrm{d}t} \mathcal{G}_{ijkl}(t) = \left[h^{(2),\mathrm{HF}}, \mathcal{G}\right]_{ijkl}(t) + \Psi^\pm_{ijkl}(t)$$

$$\mathcal{O}(N_\mathrm{t}^1)$$

- the initial values

$$G^{0,<}_{ij} = \pm \frac{1}{\mathrm{i}\hbar} n_{ij}(t_0) =: \pm \frac{1}{\mathrm{i}\hbar} n^0_{ij} \,,$$

$$\mathcal{G}^0_{ijkl} = \frac{1}{(\mathrm{i}\hbar)^2} \left\{ n^0_{ijkl} - n^0_{ik} n^0_{jl} \mp n^0_{il} n^0_{jk} \right\} \,,$$

determine the density and the pair correlations existing in the system at the initial time $t = t_0$

[10] two-particle commutator: $[A, B]_{ijkl}(t) = \sum_{pq} [A_{ijpq}(t) B_{pqkl}(t) - B_{ijpq}(t) A_{pqkl}(t)]$

The G1–G2 Scheme: beyond 2nd Born selfenergy

- other selfenergy approximations can be reformulated in the G1–G2 scheme in similar fashion:[11]

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{G}_{ijkl}(t) = \left[h^{(2),\mathrm{HF}}(t), \mathcal{G}(t)\right]_{ijkl} + \Psi^{\pm}_{ijkl}(t) + \underbrace{L_{ijkl}(t)}_{\text{TPP}} + \underbrace{P_{ijkl}(t)}_{GW} \pm \underbrace{P_{jikl}(t)}_{\text{TPH}}$$

with (times dropped)

$$L_{ijkl} := \sum_{pq}\left\{\mathfrak{h}^{L}_{ijpq}\mathcal{G}_{pqkl} - \mathcal{G}_{ijpq}\left[\mathfrak{h}^{L}_{klpq}\right]^{*}\right\}, \qquad \mathfrak{h}^{L}_{ijkl} := (\mathrm{i}\hbar)^2\sum_{pq}\left[\mathcal{G}^{\mathrm{H},>}_{ijpq} - \mathcal{G}^{\mathrm{H},<}_{ijpq}\right] w_{pqkl}\,,$$

$$P_{ijkl} := \sum_{pq}\left\{\mathfrak{h}^{\Pi}_{qjpl}\mathcal{G}_{piqk} - \mathcal{G}_{qjpl}\left[\mathfrak{h}^{\Pi}_{qkpi}\right]^{*}\right\}, \qquad \mathfrak{h}^{\Pi}_{ijkl} := \pm(\mathrm{i}\hbar)^2\sum_{pq} w^{\pm}_{qipk}\left[\mathcal{G}^{\mathrm{F},>}_{jplq} - \mathcal{G}^{\mathrm{F},<}_{jplq}\right]$$

and the Hartree/Fock (H/F) two-particle Green functions

$$\mathcal{G}^{\mathrm{H},\gtrless}_{ijkl}(t) := G^{\gtrless}_{ik}(t,t)G^{\gtrless}_{jl}(t,t)\,, \qquad \mathcal{G}^{\mathrm{F},\gtrless}_{ijkl}(t) := G^{\gtrless}_{il}(t,t)G^{\lessgtr}_{jk}(t,t)$$

- include TPP, GW and TPH terms simultaneously: dynamically-screened-ladder (DSL) approximation. Conserving, applicable to short times. No explicit selfenergy known.
- nonequilibrium generalization of ground state result (Bethe-Salpeter equation)

[11] J.-P. Joost, N. Schlünzen, and M. Bonitz, PRB **101**, 245101 (2020), Joost et al., PRB **105**, 165155 (2022);

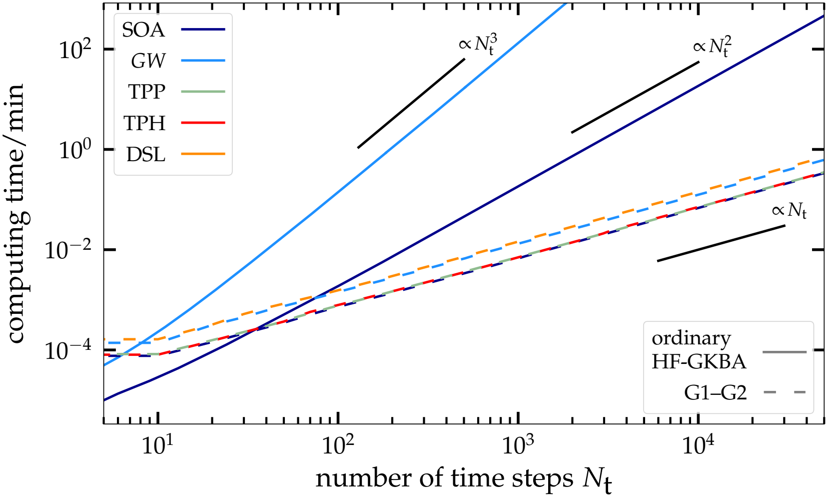
Numerical Scaling of G1–G2 vs. Standard HF-GKBA

- linear time scaling outweights introduction of 4-dimensional two-particle Green function
  → new scheme an improvement in most cases of practical relevance

		$\boldsymbol{\Sigma}$				
Basis	**HF-GKBA**	**2B**	***GW***	**TPP**	**TPH**	**DSL**
general	standard	$\mathcal{O}\left(N_\mathrm{b}^5 N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^3\right)$	–
	G1–G2	$\mathcal{O}\left(N_\mathrm{b}^5 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^6 N_\mathrm{t}^1\right)$
	speedup ratio	$\mathcal{O}\left(N_\mathrm{t}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{t}^2\right)$	–
Hubbard	standard	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	–
	G1–G2	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$
	speedup ratio	$\mathcal{O}\left(N_\mathrm{t}/N_\mathrm{b}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2/N_\mathrm{b}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2/N_\mathrm{b}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2/N_\mathrm{b}\right)$	–
HEG	standard	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^3\right)$	–
	G1–G2	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^3 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$	$\mathcal{O}\left(N_\mathrm{b}^4 N_\mathrm{t}^1\right)$
	speedup ratio	$\mathcal{O}\left(N_\mathrm{t}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2\right)$	$\mathcal{O}\left(N_\mathrm{t}^2/N_\mathrm{b}\right)$	$\mathcal{O}\left(N_\mathrm{t}^2/N_\mathrm{b}\right)$	–

Numerical Scaling of G1–G2 vs. Standard HF-GKBA

- time-linear scaling achieved quickly for all approximations. Example: 10-site Hubbard chain



Numerical G1–G2 results: TPP vs. DSL

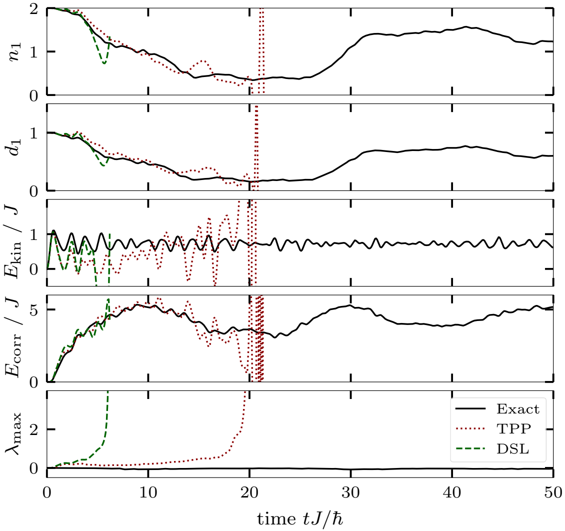


Figure 1: G1–G2 simulation for half-filled 6-site Hubbard system at moderate coupling, $U/J = 4$. Sites 1–3 are initially doubly occupied and sites 4–6 are empty. At time $t = 0$ the confinement potential is removed (quench). Instability for increasing U

G1–G2 scheme: achieving long simulation times for correlated electrons: contraction consistency and purification

- Enforcing Contraction consistency[12]:

$$\frac{N}{2} G_{ij}^{\uparrow\uparrow} = -\mathrm{i}\hbar \sum_p G_{ipjp}^{(2),\uparrow\downarrow\uparrow\downarrow}$$

$$G_{ij}^{\uparrow\uparrow} = -\mathrm{i}\hbar \sum_p G_{ippj}^{(2),\uparrow\downarrow\uparrow\downarrow}$$

$$\left(\frac{N}{2} - 1\right) G_{ijkl}^{(2),\uparrow\downarrow\uparrow\downarrow} = -\mathrm{i}\hbar \sum_p G_{ipjkpl}^{(3),\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow}$$

$$\frac{N}{2} G_{ijkl}^{(2),\uparrow\uparrow\uparrow\uparrow} = -\mathrm{i}\hbar \sum_p G_{ijpklp}^{(3),\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow}$$

$$G_{ijkl}^{(2),\uparrow\uparrow\uparrow\uparrow} = -\mathrm{i}\hbar \sum_p G_{ipjklp}^{(3),\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow}$$

$$G_{ijkl}^{(2),\uparrow\uparrow\uparrow\uparrow} = -\mathrm{i}\hbar \sum_p G_{ijpkpl}^{(3),\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow}$$

[12] see e.g. papers by Coleman, Maziotti and others
F. Lackner *et al.*, Phys. Rev. A (2015), Phys. Rev. A (2017)
J-P. Joost, N. Schlünzen, H. Ohldag, M. Bonitz, F. Lackner, and I. Brezinova, PRB 2022

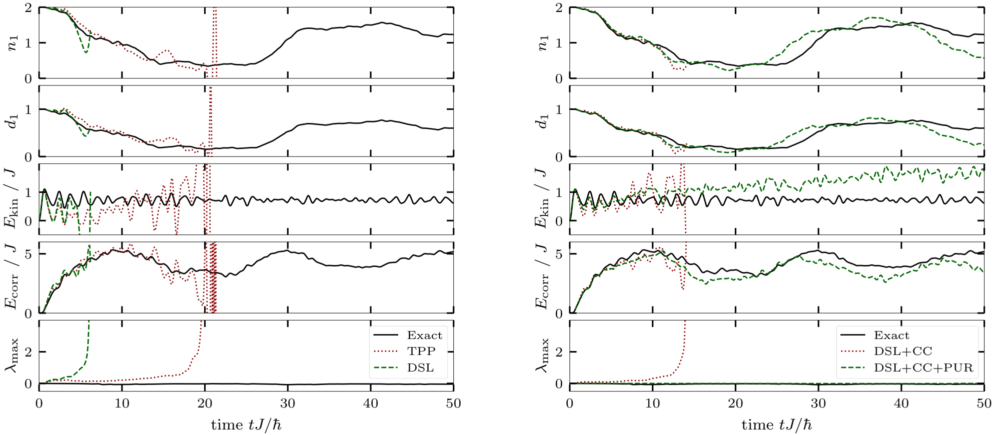


Figure 2: G1–G2 without (left) and with (right) contraction consistency (CC) and purification (PUR). Half-filled 6-site Hubbard system at moderate coupling, $U/J = 4$. Sites 1–3 are initially doubly occupied and sites 4–6 are empty. At time $t = 0$ the confinement potential is removed (quench).

[13] J-P. Joost, N. Schlünzen, H. Ohldag, M. Bonitz, F. Lackner, and I. Brezinova, PRB 2022

G1–G2 scheme: Benchmarks against DMRG, 20 electrons[14]

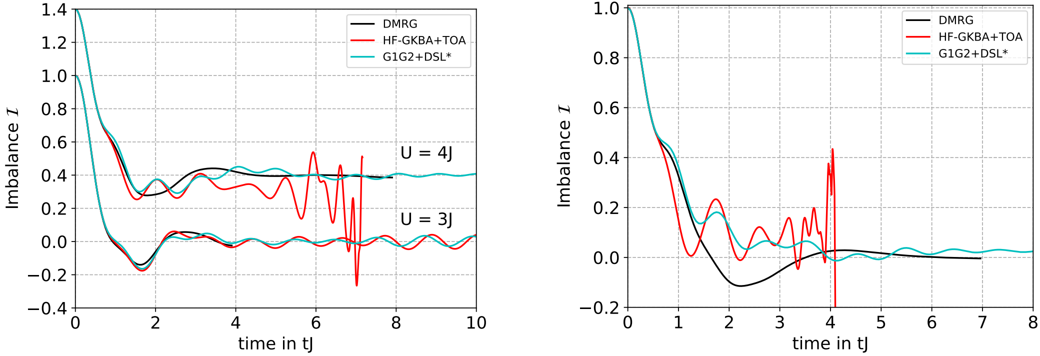


Figure 3: Relaxation of the charge Imbalance starting from a charge density wave state, $L = N = 20$ for $U/J = 3, 4$ (left) and $U/J = 5$ (right). DMRG and third order approximation (TOA) vs. G1-G2-DSL with CC and purification.

[14] J-P. Joost, N. Schlünzen, H. Ohldag, M. Bonitz, F. Lackner, and I. Brezinova, PRB 2022, DMRG and TOA data from Schluenzen et al. PRB 2017

Motivation:

1. Prediction of petahertz electronics, sub-fs space-resolved dynamics required
2. space dependent local density of states[15], site selective laser excitation and dynamics

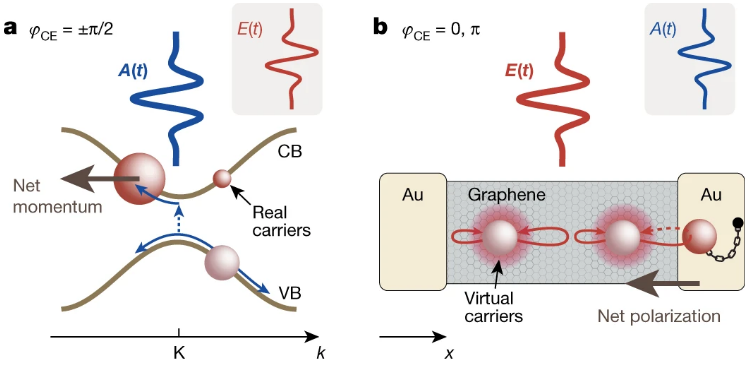
[15] J.-P. Joost, A.-P. Jauho, and M. Bonitz, Nano Letters **19**, 9045 (2019)

Predicted laser driven Petahertz electronics[16]

Experiments by P. Hommelhoff *et al.*: logic gate for lightwave electronics, variation of carrier envelope phase ϕ_{CE} of few cycle fs-laser pulse

a: momentum asymmetry $(A(t))$ creates $f_c(-k) \neq f(k)$ and net current

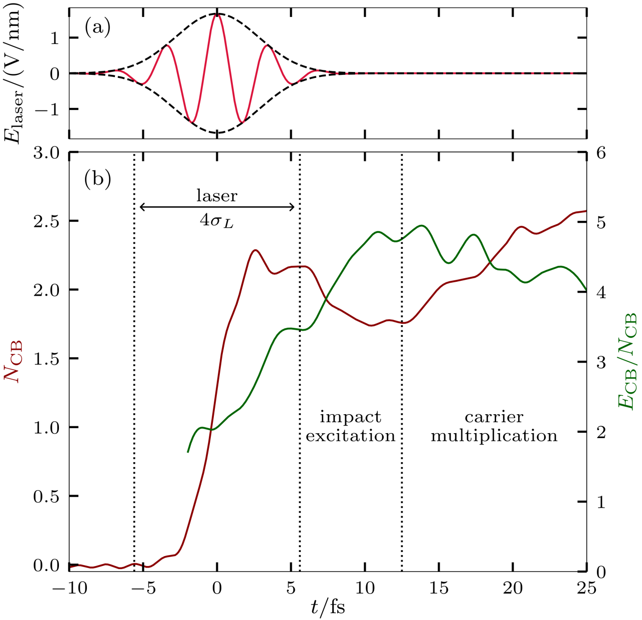
b: real space asymmetry $(E(t))$ of density creates net polarization

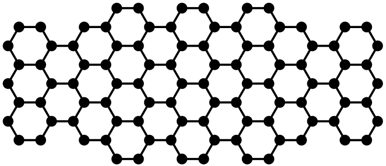


[16]Boolakee et al., Nature **605**, 251 (2022)

Short Time Carrier Dynamics

$\|, E_0 = 0.1, \omega_L = 1.2\text{eV (IR)}$

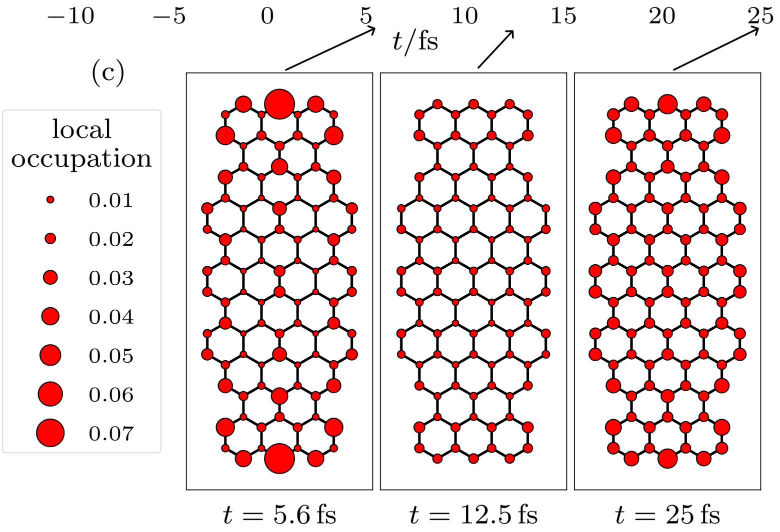




Laser parameters

- dipole approximation (wavelength μm, system nm)
- $U_{\text{pot}} = -\vec{E}_{\text{Laser}} \cdot \vec{x}$
- $E_{\text{Laser}} = E_0 \exp\left(-\frac{(t-t_0)^2}{2\sigma_L^2}\right)$
- $E_0 = 0.1$
- $\omega_L = 0.5\,J \approx 1.2\,\text{eV}$
- $\sigma_L = 10\,J^{-1} \approx 3\,\text{fs} \quad (\approx 0.2\,\text{eV})$
- polarization: parallel to ribbon ($\|$)

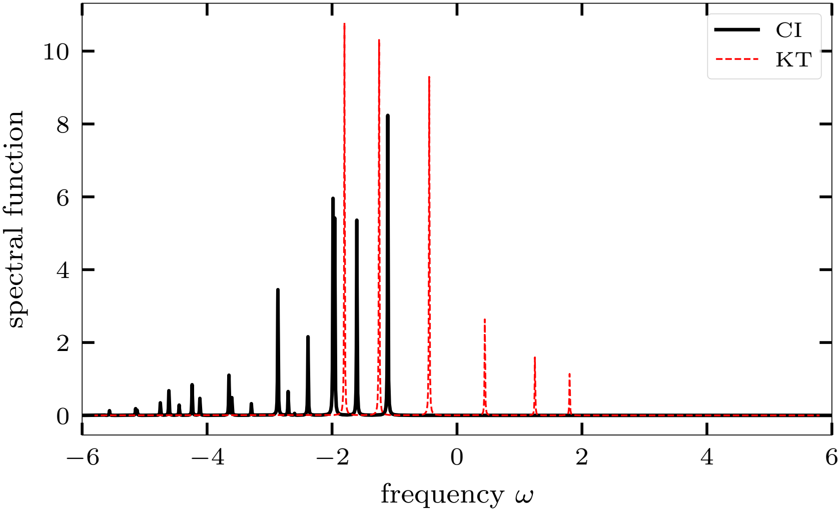
Local Occupation of Excited Electrons $\|, E_0 = 0.1, \omega_L = 1.2\text{eV (IR)}$



- excited electrons are first localized at the edges and subsequently redistributed

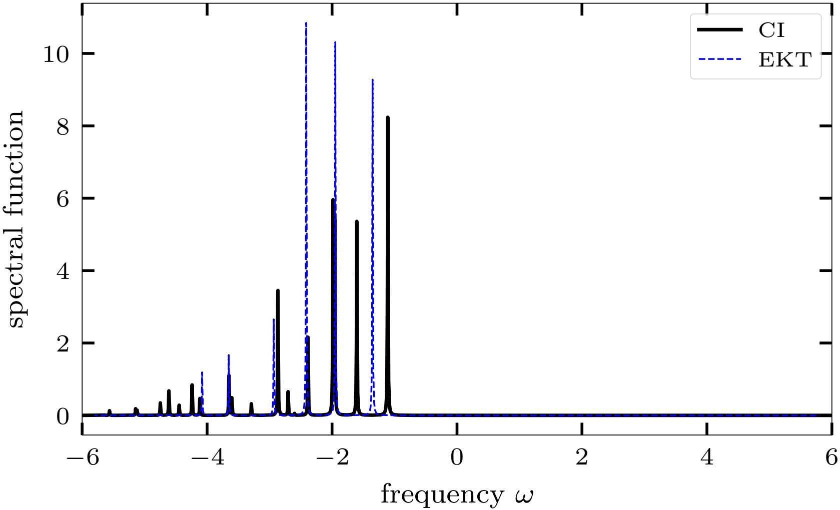
Electron spectra from G1–G2 simulations, $U/J = 4, N_B = 6$, 1D

- HF-GKBA reproduces Hartree-Fock retarded Green function, $G^R(t - t')$
- use Koopmans' theorem (ground state results)



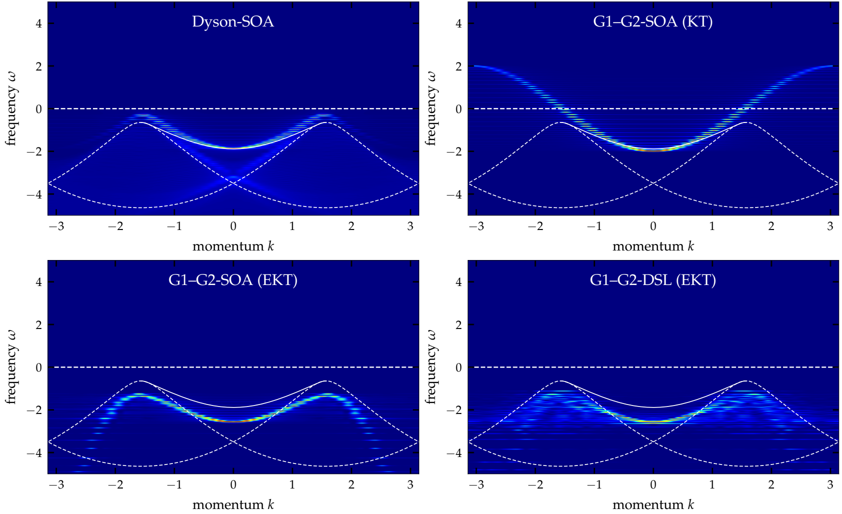
Electron spectra from G1–G2 simulations, $U/J = 4, N_B = 6$, 1D

- HF-GKBA reproduces Hartree-Fock retarded Green function, $G^R(t - t')$
- use Extended Koopmans' theorem (ground state results)



Dispersion relation from G1–G2 simulations, $U/J = 4, N_B = 54$, 1D

- HF-GKBA reproduces Hartree-Fock retarded Green function, $G^R(t - t')$
- Koopmans vs. Extended Koopmans' theorem (SOA vs. DSL), white: Bethe ansatz



approximation

Full NEGF $\longrightarrow$ HF-GKBA

$$\mathcal{O}(N_\mathrm{t}^3) \qquad\qquad \mathcal{O}(N_\mathrm{t}^2)$$

- HF-GKBA recovers Boltzmann-type kinetic equations and overcomes their problems: total energy conservation, correct short-time dynamics, correlated equilibrium state

Summary and Outlook

approximation **no approximation!**

Full NEGF → HF-GKBA → G1–G2

$\mathcal{O}(N_t^3)$ $\mathcal{O}(N_t^2)$ $\mathcal{O}(N_t^1)$

- HF-GKBA recovers Boltzmann-type kinetic equations and overcomes their problems: total energy conservation, correct short-time dynamics, correlated equilibrium state
- G1–G2 calculations can be done in linear time[17]
- in most cases this results in significant speed-ups ($\times 10^2$–10^4, despite rank-4 $\mathcal{G}$)
- Full nonequilibrium DSL (combining dyn screening and strong coupling) simulations possible
- **Price to pay:** expensive storage of $\mathcal{G}_{ijkl}(t) \rightarrow$ test alternative representation

[17] N. Schlünzen, J.-P. Joost and M. Bonitz, Phys. Rev. Lett. **124**, 076601 (2020)
J.-P. Joost, N. Schlünzen, and M. Bonitz, Phys. Rev. B **101**, 245101 (2020)
J.-P. Joost, N. Schlünzen, H. Ohldag, M. Bonitz, F. Lackner, and I. Brezinova, Phys. Rev. B **105**, 165155 (2022)

Quantum Fluctuations: Definitions[18]

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Expectation values and fluctuations of Green functions: $\hat{G} = G + \delta\hat{G}$

$$G^{>}_{ij}(t) = \frac{1}{\mathrm{i}\hbar}\langle \hat{a}_i(t)\hat{a}^{\dagger}_j(t)\rangle, \qquad \hat{G}^{>}_{ij}(t) = \frac{1}{\mathrm{i}\hbar}\,\hat{a}_i(t)\hat{a}^{\dagger}_j(t)$$

$$G^{<}_{ij}(t) = \pm\frac{1}{\mathrm{i}\hbar}\,\langle \hat{a}^{\dagger}_j(t)\hat{a}_i(t)\rangle, \qquad \hat{G}^{<}_{ij}(t) = \pm\frac{1}{\mathrm{i}\hbar}\,\hat{a}^{\dagger}_j(t)\hat{a}_i(t)$$

$$\delta\hat{G}_{ij}(t) := \delta\hat{G}^{<}_{ij}(t) = \delta\hat{G}^{>}_{ij}(t)$$

Extension to N-particle fluctuations:

$$\Gamma^{(N)}_{i_1 i_2 \dots i_N; j_1 j_2 \dots j_N}(t) := \langle \delta\hat{G}_{i_1 j_1}(t)\delta\hat{G}_{i_2 j_2}(t)\dots\delta\hat{G}_{i_N j_N}(t)\rangle$$

Short notations:

$$\gamma_{ij;kl}(t) := \Gamma^{(2)}_{ij;kl}(t)\,, \qquad \Gamma_{ijk;lmn}(t) := \Gamma^{(3)}_{ijk;lmn}(t)$$

[18] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022); arXiv:2204.08250

Exact single-particle dynamics:

Collision integral in terms of fluctuations (instead of $\mathcal{G}$)

$$\mathrm{i}\hbar\partial_t G^<_{ij}(t) = \left[h^{\mathrm{H}}, G^<\right]_{ij}(t) + \left[\tilde{I} + \tilde{I}^\dagger\right]_{ij}(t)$$

$$h^{\mathrm{H}}_{ij}(t) = h_{ij}(t) + U^{\mathrm{H}}_{ij}(t)\,; \quad U^{\mathrm{H}}_{ij}(t) = \pm\mathrm{i}\hbar\sum_{kl} w_{ikjl}(t)G^<_{lk}(t)$$

$$\left[\tilde{I} + \tilde{I}^\dagger\right]_{ij}(t) = \pm\mathrm{i}\hbar\sum_{klp}\left\{w_{iklp}(t)\gamma_{plkj}(t) - w_{kljp}(t)\gamma_{ipkl}(t)\right\}$$

Eliminate dynamics of $\gamma_{ij;kl}(t) = \langle\delta\hat{G}_{ik}(t)\delta\hat{G}_{jl}(t)\rangle$ by propagating $\delta\hat{G}(t)$:

Exact equation for single-particle fluctuation (two-point function)

$$\mathrm{i}\hbar\partial_t\delta\hat{G}_{ij}(t) = \left[h^{\mathrm{H}}, \delta\hat{G}\right]_{ij}(t) + \left[\delta\hat{U}^{\mathrm{H}}, G^<\right]_{ij}(t) + \left[\delta\hat{U}^{\mathrm{H}}, \delta\hat{G}\right]_{ij}(t) - \left[\tilde{I} + \tilde{I}^\dagger\right]_{ij}(t)$$

$$\delta\hat{U}^{\mathrm{H}}_{ij}(t) = \pm\mathrm{i}\hbar\sum_{kl} w_{ikjl}(t)\delta\hat{G}^<_{lk}(t)$$

[19] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022); arXiv:2204.08250

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Stochastic Mean field idea:[20]

- replace quantum-mechanical expectation value by semiclassical mean over realizations A^λ,

$$\langle \hat{A} \rangle \longrightarrow \overline{A^\lambda}$$

- Random sampling of initial conditions of non-interacting system:

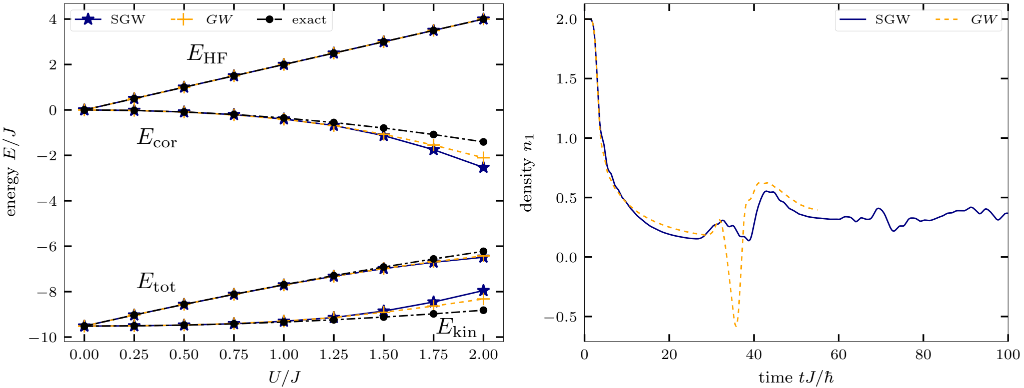
$$\begin{aligned} \overline{\Delta G^\lambda_{ij}(t_0)} &= 0, \\ \overline{\Delta G^\lambda_{ik}(t_0)\Delta G^\lambda_{jl}(t_0)} &= -\frac{1}{2\hbar^2}\delta_{il}\delta_{jk}\{n_i(1 \pm n_j) + n_j(1 \pm n_i)\}. \end{aligned}$$

- Careful test of probability distribution and sampling methods[21]
- interactions turned on via adiabatic switching
- Stochastic polarization approximation reproduces time-dependent GW–G1–G2 results[22]

[20] S. Ayik, Phys. Lett. B **658**, 174 (2008), D. Lacroix, S. Hermanns, C. M. Hinz, and M. Bonitz, PRB **90**, 125112 (2014)

[21] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022); arXiv:2204.08250

Quantum Fluctuations: Testing the stochastic GW-approximation[22]



Left: Ground state for $N_s = N = 8$. **Right:** Density dynamics following confinement quench for $N_s = 30$, $N = 10$, $\nu = 1/6$, $U = 1$. Initially leftmost 5 sites doubly occupied, rest empty.

[22] E. Schroedter, J.-P. Joost, and M. Bonitz, Cond. Matt. Phys. **25**, 23401 (2022); arXiv:2204.08250

Scaling of selected quantum many-body methods revisited

G1–G2 scheme allows for highly efficient accurate, long and stable quantum dynamics, for systems of any geometry and time scale